

Double exchange model on triangular lattice: non-coplanar spin configuration and phase separation near quarter filling

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Abstract. The effect of antiferromagnetic (AF) superexchange (SE) interaction on double exchange model in two-dimensional triangular lattice is investigated in this Letter. The long-range ferromagnetic (FM) spin-spin correlation and strong electron-spin correlation in triangular lattice are two important factors for truncation error of FM-like spin-spin correlation near quarter filling by truncated polynomial expansion, which can be overcome by high expansion moment M . Near quarter filling, non-coplanar spin configuration is induced by competition between FM parallel alignment mediated by itinerant electrons and AF SE interaction between localized spins. A phase transition from FM to non-coplanar phase is characterized by both spin structure factor and the change of the slope of energy in term of AF SE interaction, which is accompanied by a phase separation in electronic density. At strong AF SE interaction, there is also another phase separation accompanying the phase transition from non-coplanar spin phase to 120-degree spin phase. Finally we discuss the effect of temperature on magnetic structure.

AMS subject classifications: 82B05, 26C99, 65F99, 15A30

Key words: Manganite, non-coplanar spin configuration, phase separation, frustration, triangular lattice, Monte Carlo simulation, polynomial moment expansion.

1 Introduction

Recently noncoplanar spin configuration emerging in the ferromagnetic (FM) Kondo model becomes one of research hot topics in strongly correlated electrons field. It is induced by the competition between FM alignment of localized spins from Hund-interaction

of itinerant electron and antiferromagnetic (AF) superexchange (SE) interaction on regular lattices and frustrated lattices [1–3]. In Ref. [1], this kind of noncoplanar spin configuration was verified by the average of spin-spin correlation and the inner product of three localized spins located at one triangle in triangular lattice at quarter filling via classic Monte Carlo simulation combined with truncated polynomial expansion method (TPEM) [4, 5]. This kind of peculiar spin configuration is stable in quite large parameters space [3]. 120-degree spin configuration is easily destabilized by electron doping from band bottom and a noncoplanar three-sublattice ordering occurs accompanied by an intervening phase separation, while a noncoplanar ordering does not occur by hole doping from band top [3]. Phase separation [6, 7] is a key factor for colossal magnetoresistance effect (CMR) [8, 9]. In lightly doped region, there exists a phase separation from the electronic density [3].

Geometrical frustration and/or magnetic frustration from competition between FM double exchange (DE) interaction and AF SE interaction arouse a broad interest, inspired by the discovery of peculiar transport properties, such as unconventional anomalous Hall effect, in some pyrochlore oxides [10–12]. In Ref. [2], the density of states and the resistivity at various J_{AF} and temperature were investigated at quarter filling. In noncoplanar spin configuration, they are independent of the temperature. The system undergoes a metal-insulator transition when J_{AF} increases. The bad metallic behavior is consistent with experimental observation in frustrated itinerant magnets $R_2\text{Mo}_2\text{O}_7$ [2].

In Ref. [1], we had studied the spin-spin correlation and spin structure factor at various electron-filling cases. However, in this Letter, we focus on the spin configuration around quarter filling. As shown in Ref. [1], at quarter filling in triangular lattice, accurate FM-like spin-spin correlation can only be reproduced with higher moment M of polynomial expansion, e.g. $M \simeq 200$. Here we find that the long-range FM-like correlation between spin-spin correlation is responsible for truncation error of TPEM, by varying the dimension of systems. However, FM-like spin-spin correlation at low electron filling region, e.g. $n \simeq 0.1944$, can be accurately reproduced as $M \simeq 30$, as shown in Fig. 4(c) [1]. This fact implies that electron-spin correlation in FM Kondo model is also important for truncation error of TPEM. In contrast to large truncation error of FM-like spin-spin correlation near quarter filling at low truncation moment M , most physical quantities, such as the energy E and the electron density n , are accurate at various parameters.

We investigate the effect of AF SE interaction on the spin configuration near quarter filling. Non-coplanar spin configuration is induced by competition between FM parallel alignment mediated by itinerant electrons and AF SE interaction between localized spins. A phase transition from FM to non-coplanar phase is characterized by both spin structure factor and the change of the slope of energy in term of AF SE interaction. It is accompanied by a phase separation in electronic density. There is also another phase separation accompanying the phase transition from non-coplanar spin phase to 120-degree spin phase at strong AF SE interaction. It was found that a phase separation also occurs in lightly doped case in Ref. [3]. Finally we discuss the effect of temperature on magnetic structure.

2 Hamiltonian, triangular lattice and spin structure factor

The hamiltonian consists of two parts, i.e., $H = H_{DE} + H_{AF}$. Double-exchange model H_{DE} describes the Hund's-rule interaction between itinerant electron and localized spins,

$$H_{DE} = -t \sum_{\langle ij \rangle, \alpha} (C_{i,\alpha}^\dagger C_{j,\alpha} + h.c.) - J_H \sum_{i,\alpha,\beta} C_{i,\alpha}^\dagger \sigma_{\alpha\beta} C_{i,\beta} \cdot S_i. \quad (2.1)$$

Here the nearest-neighbor hopping integral t is adopted as the energy unit and J_H is the Hund interaction strength. $C_{i,\alpha}^\dagger$ ($C_{i,\alpha}$) creates (annihilates) one electron at site i with spin α , $\langle ij \rangle$ stands for the nearest neighbors of lattice site, and $\sigma_{\alpha\beta}$ is the Pauli matrix. The localized spin S_i at site i is assumed as 1 here. $H_{AF} = J_{AF} \sum_{\langle ij \rangle} S_i \cdot S_j$ describes AF SE interaction between nearest-neighboring lattice sites. Localized spins is treated as a classic field ϕ and electron degree of freedom can be integrated for any given localized spin configuration. Classic Monte Carlo simulation combined with TPME [4, 5] is adopted to simulate the energy, electron density, spin-spin correlation and spin structure factor. There are three key parameters for TPME, i.e., M , ϵ_p and ϵ_{tr} , which control the accuracy and computational speed [1, 4, 5]. The maximal Monte Carlo step is 40000, and the physical quantity is evaluated every 20 steps after first 6000 warmup steps.

6×6 triangular lattice is shown in Fig. 1(a) and the lattice constant is denoted as a . Periodic boundary condition is adopted in simulation. Each lattice site has six nearest neighboring lattice sites. Two basic lattice vectors are $\vec{a}_1 = (1, 0)a$ and $\vec{a}_2 = (\frac{1}{2}, \frac{\sqrt{3}}{2})a$, as shown in Fig. 1(b). The reciprocal lattices are $\vec{b}_1 = \frac{4\pi}{\sqrt{3}a}(\frac{\sqrt{3}}{2}, -\frac{1}{2})$ and $\vec{b}_2 = \frac{4\pi}{\sqrt{3}a}(0, 1)$, and the first Brillouin zone is also shown in Fig. 1(c). For $L \times L$ triangular lattice, similar to Ref. [1], the momentum $\vec{q} = \frac{m}{L}\vec{b}_1 + \frac{n}{L}\vec{b}_2$ is shortened as (q_1, q_2) , with $q_1 = \frac{m}{L}$, $q_2 = \frac{n}{L}$, and m, n as integers from 0 to L . Spin structure factor $S(q)$ is defined by

$$S(\vec{q}) = \frac{1}{N} \sum_{i,j} \langle S_i \cdot S_j \rangle e^{i\vec{q} \cdot \vec{r}_{ij}}, \quad (2.2)$$

where $\langle S_i \cdot S_j \rangle$ and \vec{r}_{ij} is the average spin-spin correlation and the displacement between site i and j respectively, and $N = L \times L$. Here $j = i_x + (i_y - 1) * L$ is the ordering of lattice site with indices (i_x, i_y) , in which $1 \leq i_x \leq L$ and $1 \leq i_y \leq L$. Due to periodic boundary condition, only $\langle S_1 \cdot S_j \rangle$ is shown.

3 Result and discussion

3.1 The truncation error of TPME related to the range of FM spin-spin correlation and electron-spin correlation

It is well known that the truncation error decreases when the truncation moment M of TPME increases. Therefore the physical quantity can be exactly reproduced when M

approaches infinity. As discussed in Ref. [1], a large truncation moment ($M \geq 200$) is required to reproduce FM-type spin-spin correlation $\langle S_1 \cdot S_j \rangle$ in 2D triangular lattice at quarter-filling case, as shown in Fig. 2(f). Since there is no stable FM phase for double exchange model in these systems with periodic boundary condition at quarter filling, FM spin-spin correlation is short-ranged. Therefore, $M \simeq 30$ and $M \simeq 50$ is enough to reproduce the spin-spin correlation $\langle S_1 \cdot S_j \rangle$ at quarter filling in 1D and 2D regular lattice respectively, as shown in Fig. 2(a,c). At strong AF SE interaction, e.g., $J_{AF} = 0.1$, spin-spin correlation is FM and even AF in short range, $M = 30$ is enough as shown in Fig. 2(b,d) and Fig. 4(e) in Ref. [1]. For 2D square lattice, a spin-flux phase [13,14] occurs, where four localized spins within each square lie (anti)clockwise at the same plane. For 2D triangular lattice, a new type of spin-flux phase, i.e., noncoplanar spin configuration, occurs as a result of competition of electron-mediated FM alignment and AF SE interaction [1–3]. These results show that the range of FM-type spin-spin correlation and thus FM magnetic domain is an important factor for the accuracy of spin-spin correlation by TPME.

On the other hand, we explore the effect of electron mediation on the accuracy of spin-spin correlation by TPME. In three-dimensional (3D) cube lattices, the magnetization at quarter filling is accurate at $M = 30$ [5], compared with exact diagonalization method. Though FM-type spin-spin correlation at low electron filling region (e.g., $n=0.1944$) is long-ranged in 2D triangular lattice, FM-type spin-spin correlation is accurate at $M = 30$, as shown in Fig. 4(c) [1]. The electron spin correlations in both cases in above are weaker than that in triangular lattice near quarter filling, therefore electron mediation and electron-spin correlation is the other important factor for the accuracy of spin-spin correlation by TPME.

Compared with spin-spin correlation, the energy can be reproduced by TPME at $M \simeq 50$, irrelevant of the strength of AF SE interaction J_{AF} , as shown in Fig. 2(f). Therefore most of physical properties are correct by TPME, even if long-range FM spin-spin correlation in FM phase is overestimated. We notice that the energy first increases, reaches a maximum and then decreases, as J_{AF} increases from 0 to 0.1, which indicates a phase transition from FM into non-coplanar spin configuration, to be discussed in the following.

3.2 Effect of AF superexchange interaction on spin configuration

Due to simplicity, one triangular lattice is chosen to show the spin configuration under various AF superexchange interaction. The possible electron density n_e of one triangular lattice is 0, 1/3, 2/3 and 1. The spins are parallel at $n_e = 1/3$ at weak AF SE interaction, while they are in 120° at $n_e = 2/3$ and 1, independent of the value of J_{AF} . When J_{AF} is 0.17~0.18, the electron density sharply changes from 1/3 to 2/3, which indicates a phase separation, as shown in Fig. 3(a). The energy E first increases, reaches a maximum -0.49071 and then monotonically decreases, when J_{AF} increases from 0 to 0.4, as shown in Fig. 3(b). Interestingly, the maximal energy E_{max} occurs in the vicinity of the phase separation. Meanwhile the spin-spin correlation changes from 1 to -0.5, when J_{AF}

increases from 0 to 0.4, as shown in Fig. 3(c). The spin configuration evolves from parallel to non-coplanar configuration to 120° . The probability distribution of the inner product of the nearest-neighboring localized spins $S_1 \cdot S_2$ is analyzed at $J_{AF} = 0.1, 0.17$ and 0.4 , as shown in Fig. 3(d). The mean value of $\langle S_1 \cdot S_2 \rangle$ is $0.8381, 0.1753$ and -0.48842 respectively. It clearly shows that the peak of probability distribution is located at $1, -0.5$ for FM phase and 120° . However, the probability distribution has two peaks at 1 and -0.5 respectively for non-coplanar spin configuration. We stress that the phase separation in one triangular lattice is different from those in large triangular lattice, since $n_e = 2/3$ induces 120° spin configuration. Therefore non-coplanar spin configuration in one triangular lattice exists in a small space of parameters.

Here we investigate the effect of AF superexchange interaction on the energy E , the electron density n , the spin-spin correlation $\langle S_i \cdot S_j \rangle$ and spin structure factor $S(\vec{q})$, by continuously tuning the strength of AF superexchange interaction J_{AF} . As J_{AF} increases from 0 to $0.038t$, the energy E increases, as shown in Fig. 4(a). At small J_{AF} , the spin configuration is in FM phase and $\langle S_i \cdot S_j \rangle$ is positive. The FM phase is strengthened by the six nearest neighboring lattice sites in 2D triangular lattice [1], compared with unstable FM phase in 2D square lattice. When J_{AF} increases, FM-like spin-spin correlation decreases and the angle between nearest neighboring localized spins approaches to 90° , as shown in Fig. 4(b). As a result, the spin structure factor at the vector $(0,0)$, i.e. $S(0,0)$, decreases as J_{AF} increases, as shown in Fig. 4(c). Since $\sum_{\langle ij \rangle} \langle S_i \cdot S_j \rangle$ remains positive, the energy E increases with J_{AF} by a competition between increasing J_{AF} and decreasing $\langle S_i \cdot S_j \rangle$.

As J_{AF} is larger than a critical value $J_{AF}^{c1} = 0.038t$, the energy E decreases from a maximum. Therefore the slope of the energy in term of J_{AF} changes sign from positive to negative, as shown in Fig. 4(a). The change in the slope of the energy usually indicates a phase transition [15]. Interestingly, a phase separation occurs around this phase transition, which is indicated by the electron density, as shown in Fig. 4(a). As $J_{AF} > J_{AF}^{c1}$, the energy corresponding to FM phase is too large and a new phase with a lower energy emerges, i.e., noncoplanar spin configuration. In this phase, the spin-spin correlation between any two nearest neighboring localized spins is negative, as shown in Fig. 4(b), while $(S_i \times S_j) \cdot S_k$ with the sites i, j and k belonging to one triangle is nonzero [1]. Noncoplanar spin configuration is different from both FM phase and 120° configuration phase. The spin structure factor $S(\vec{q})$ has three peaks, which is located at vectors $(0.5,0)$, $(0,0.5)$ and $(0.5,0.5)$ respectively, as shown in Fig. 4(c), which is consistent with Ref. [2]. Rescaled spin structure factor $S(\vec{q})/N \sim 0.29$ is close to its maximum value $1/3$, and finite size effect is small [1].

When $J_{AF} > J_{AF}^{c2} = 0.225t$, the system eventually evolves into 120° spin configuration phase. In this phase, the spin-spin correlation between any two nearest neighboring localized spins closes to -0.5 , as shown in corrected Fig.4(a,d) [1], while $(S_i \times S_j) \cdot S_k$ with the sites i, j and k belonging to one triangle is zero [1]. The spin structure factor $S(\vec{q})$ has two peaks, which is located at vectors $(1/3, 2/3)$ and $(2/3, 1/3)$ respectively, as shown in corrected Fig. 5(a,d) [1, 16], which is consistent with Ref. [2]. Rescaled spin structure factor $S(\vec{q})/N$ is close to its maximum value $1/2$ [1, 2]. There is another phase separation

at $J_{AF}^2 = 0.225t$ indicated by the electron density. Interestingly, this phase separation indicates phase transition from noncoplanar spin configuration to 120-degree spin phase, which is consistent with the phase diagram shown in Ref. [2] in the limit of infinite Hund interaction.

3.3 Effect of temperature on spin configuration

We further investigate the effect of temperature on the spin-spin correlation at $J_{AF} = 0$ and 0.1 respectively, as shown in Fig. 5(a,b). As the temperature T increases from 0 to $0.1t$, FM-type spin-spin correlation at $J_{AF} = 0$ decreases as shown in Fig. 5(a), and the spin configuration tends to be paramagnetic at high temperature. On the other hand, the absolute value of the spin-spin correlation in non-coplanar spin configuration also decreases as T increases, as shown in Fig. 5(b). When J_{AF} increases from 0 to 0.1 , the peak of spin structure factor moves from vector $(0,0)$ to vectors $(0,0.5)$, $(0.5,0)$ and $(0.5,0.5)$. Meanwhile $S(0,0)$ decreases and $S(0,0.5)$ increases. The rescaled peak of spin structure factor $S(q_x, q_y)$ decreases when the temperature T increases, as shown in Fig. 5(c). All rescaled peaks collapse at $T = 0.1$, which implies the system is in paramagnetic phase. The dependence of spin-spin correlation on temperature and no phase transition at high temperature is consistent with the phase diagram shown in Ref. [2].

4 Conclusion

We investigate the effect of AF SE interaction on the spin configuration near quarter filling and discuss the origin of the truncation error of long-range FM-like spin-spin correlation by truncated polynomial expansion. Non-coplanar spin configuration is induced by competition between FM parallel alignment mediated by itinerant electrons and AF SE interaction between localized spins. A phase transition from FM to non-coplanar phase is characterized by both spin structure factor and the change of the slope of energy in term of AF SE. It is accompanied by a phase separation in electronic density. At strong AF SE interaction, there is also another phase separation accompanying the phase transition from non-coplanar spin phase to 120-degree spin phase. Finally we discuss the effect of temperature on magnetic structure.

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- [16] G. P. Zhang, Erratum to Commun. Comput. Phys. 10, 422-432 (2011), the peaks of spin structure factor should be located at $(1/3, 2/3)$ and $(2/3, 1/3)$.

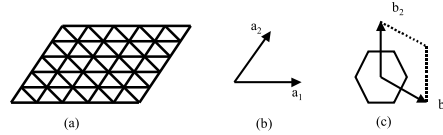


Figure 1: 6×6 triangular lattice (a), lattice vector (b), and reciprocal lattice and the first Brillouin zone (c).

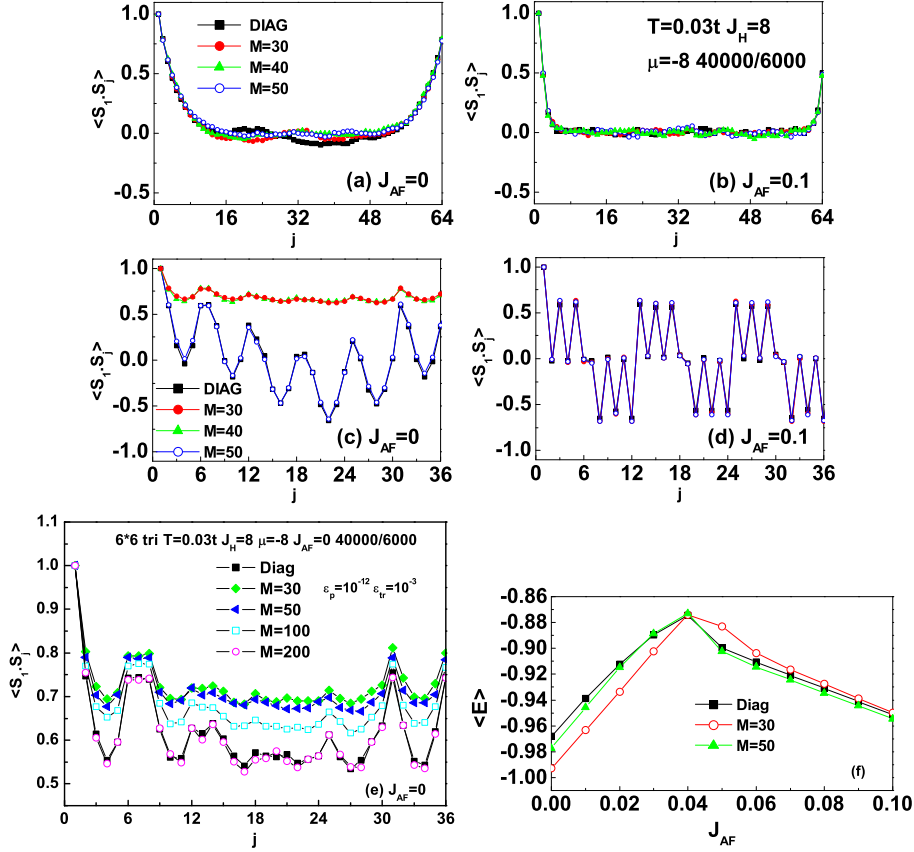


Figure 2: The comparison of spin-spin correlation $\langle S_1 \cdot S_j \rangle$ between exact diagonalization (ED) and TPEM in (a,b) one-dimensional chain with $L=64$, (c,d) two-dimensional 6×6 square lattices and (e) 6×6 triangular lattice. $J_{AF}=0$ in (a,c,e) and $J_{AF}=0.1$ in (b,d). The case for $J_{AF}=0.1$ in 6×6 triangular lattice had been shown in Fig. 4(e) [1]. (f) The relation of the energy E to the strength of antiferromagnetic superexchange interaction J_{AF} calculated by ED and TPEM with $M=30$ and $M=50$. $T=0.03t$, $J_H=8$, $\mu=-8$, $\epsilon_p=10^{-5}$ and $\epsilon_{tr}=10^{-3}$. (e) $\epsilon_p=10^{-12}$.

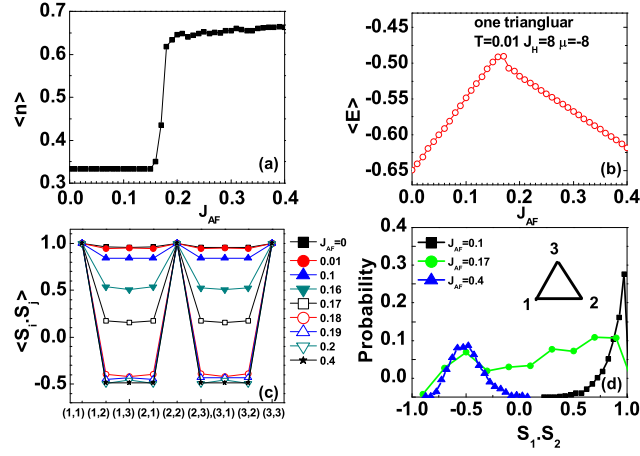


Figure 3: The electron density n (a), the energy E (b), the spin-spin correlation $\langle S_i \cdot S_j \rangle$ (c), and the probability distribution of $S_1 \cdot S_2$ (d) at various J_{AF} . The structure is one triangular lattice. Other parameters are $T=0.01t$, $J_H=8$ and $\mu=-8$.

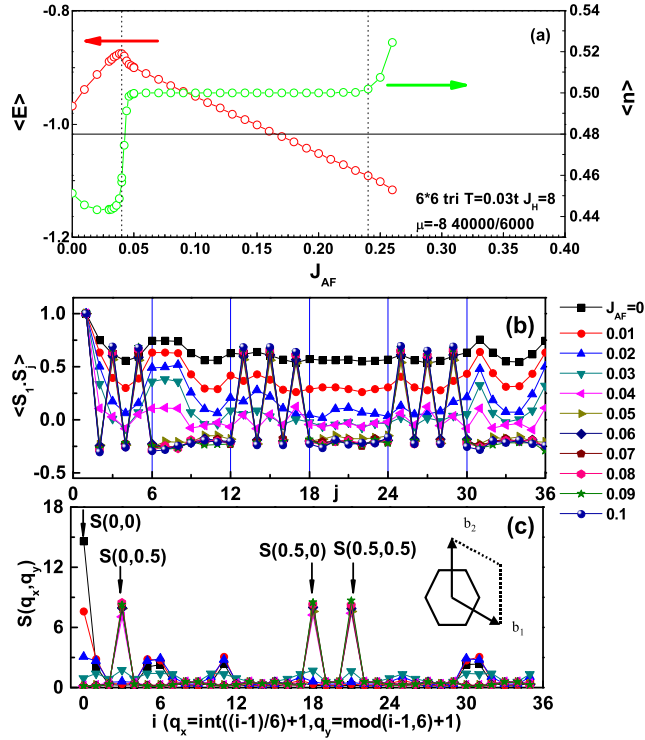


Figure 4: The energy E and the electron density n (a), the spin-spin correlation $\langle S_i \cdot S_j \rangle$ (b), and the spin structure factor $S(q_x, q_y)$ (c) when J_{AF} changes from 0 to 0.1. $T=0.03t$, $J_H=8$ and $\mu=-8$.

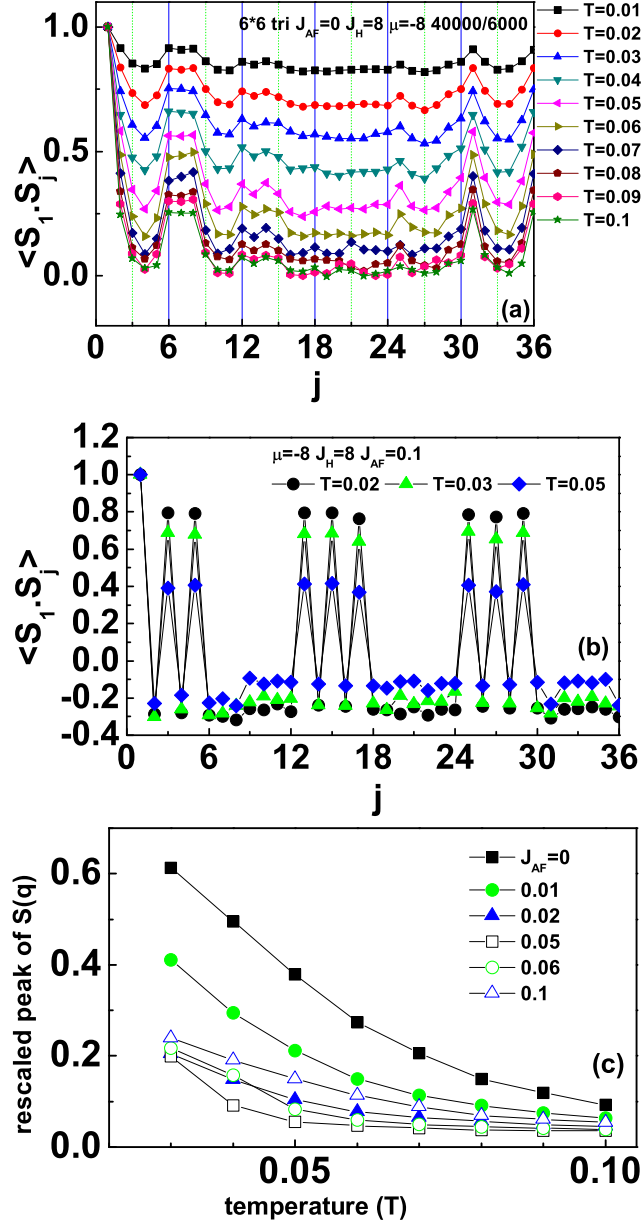


Figure 5: The dependence of spin-spin correlation (a) $\langle S_1 \cdot S_j \rangle$ at $J_{AF}=0$ and (b) $J_{AF}=0.1$ on the temperature, and (c) the dependence of rescaled peak of $S(q_{x,q_y})$ on the temperature.